Note on MacLennan-Zubarev Ensembles and QuasiStatic Processes

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Relation between natural nonequilibrium steady states of Ruelle and MacLennan-Zubarev ensembles is discussed for a system consisting of M infinitely extended systems coupled with a small system. And a characterization of quasistatic processes is investigated for a small system coupled with a single reservoir.

I. INTRODUCTION

The understanding of irreversible phenomena including nonequilibrium steady states is a long-standing problem of statistical mechanics. Recent progress in the research of mesoscopic systems brings a new aspect into this problem. In these systems, coherence (a quantum dynamical aspect) may be observed in a dissipative transport (an irreversible phenomenon) and the two aspects should be discussed simultaneously. Usually, a mesoscopic system couples with much larger environments and the interaction is not weak. As a result, the system cannot be clearly distinguished from the environments. Therefore, it is natural to deal with a mesoscopic system plus its environments as an infinitely extended system.

Statistical mechanics of infinitely extended systems has been developed so far [1, 2, 3] and, recently, their nonequilibrium properties are studied intensively. Those include analytical studies on nonequilibrium steady states of harmonic crystals [4, 5], a one-dimensional gas [6], unharmonic chains [7], an isotropic XY-chain [8], systems with asymptotic abelianness [9], a one-dimensional quantum conductor [10], an interacting fermion-spin system [11], fermionic junction systems [12], a quasi-spin model of superconductors [13] and a bosonic junction system with Bose-Einstein condensates [14]. Entropy production has been rigorously studied as well (see Refs. [9, 11, 15, 16, 17, 18], and the references therein). See also reviews [19, 20].

For C*-dynamical systems with L^1-asymptotic abelian properties, Ruelle[9] showed the existence and stability of nonequilibrium steady states (NESS) which are naturally obtained from local equilibrium ensembles. We previously announced[20] that, under stronger conditions, these natural NESS can be regarded as nonequilibrium ensembles proposed by MacLennan[21] and Zubarev[22]. In this article, we give the proof of this statement. On the other hand, one of us (ST) investigated[23] a thermodynamic behavior of a driven small system coupled with an infinitely extended reservoir. Another purpose of this article is to give the detail proof and generalization of this observation. The rest part is arranged as follows: In the next section, we summarize the setting. In Sec. III, the relation between natural NESS and Maclennan-Zubarev ensembles is discussed. Sec. IV is devoted to the discussions of the thermodynamic bahavior of a small system coupled with an infinitely extended reservoir. Some remarks are given in Sec. V.

II. C*-DYNAMICAL SYSTEMS

A. Field algebra

The system $\mathcal{S}$ in question is described by a collection $\mathcal{F}$ of all finite observables called a C*-algebra, which is a complete linear space with a norm $\|\cdot\|$, where a procduct AB and antilinear involution *: $A \to A^*$ ($\forall A, B \in \mathcal{F}$) are defined and whose norm satisfies $\|AB\| \leq \|A\|\|B\|$ and the C*-property: $\|A^*A\| = \|A\|^2$. We consider the case where an autonomous time evolution is defined which is described by a strongly continuous one-parameter group of *-automorphisms τ_t ($t \in \mathbf{R}$). Namely, τ_t is a linear map satisfying $\tau_t(AB) = \tau_t(A)\tau_t(B)$, $\tau_t(A^*) = \tau_t(A)^*$, $\tau_0 = I$ (I: the identity map), $\tau_t\tau_s = \tau_{t+s}$ and $\lim_{t\to 0}\|\tau_t(A) - A\| = 0$ ($\forall A \in \mathcal{F}$). Then, according to the theory of semigroups, there exists a densely defined generator δ of τ_t:

$$\lim_{t\to 0}\left\|\delta(A) - \frac{1}{t}\{\tau_t(A) - A\}\right\| = 0 \ , \quad (\forall A \in D(\delta))$$

where $D(\delta)$ is the domain of δ.

We further assume that two more *-automorphisms are defined:

(i) a strongly continuous L-parameter group of *-automorphisms $\alpha_{\vec{\varphi}}$ ($\vec{\varphi} \in \mathbf{R}^L$) satisfying $\alpha_{\vec{\varphi}_1}\alpha_{\vec{\varphi}_2} = \alpha_{\vec{\varphi}_1 + \vec{\varphi}_2}$, which represents the gauge transformation.

(ii) an involutive *-automorphism Θ which is represented as $\Theta = \alpha_{\vec{\varphi}_0}$ with some $\vec{\varphi}_0 \in \mathbf{R}^L$.

The automorphisms τ_t, $\alpha_{\vec{\varphi}}$ and Θ are assumed to commute with each other:

$$\Theta\tau_t = \tau_t\Theta, \quad \Theta\alpha_{\vec{\varphi}} = \alpha_{\vec{\varphi}}\Theta, \quad \tau_t\alpha_{\vec{\varphi}} = \alpha_{\vec{\varphi}}\tau_t \quad (\forall t \in \mathbf{R}, \forall \vec{\varphi} \in \mathbf{R}^L) \ .$$

A subalgebra $\mathcal{A} \subset \mathcal{F}$ consisting of invariant elements under the gauge transformations $\alpha_{\vec{\varphi}}$ ($\vec{\varphi} \in \mathbf{R}^L$) is called the observable algebra, which describes observable physical quantities. The *-automorphism Θ defines the even and odd subalgebras, respectively, $\mathcal{F}_+$ and $\mathcal{F}_-$:

$$\mathcal{F}_{\pm} = \{A \in \mathcal{F}; \Theta(A) = \pm A\} \ .$$

When the system involves fermions, even and odd subalgebras correspond to dynamical variables which are sums of products of, respectively, even and odd numbers of fermion creation and/or annihilation operators. Note that, since $\alpha_{s\vec{e}_\lambda}$ ($s \in \mathbf{R}$, $\vec{e}_\lambda$: a unit vector whose λth element is unity) defines a strongly continuous group of *-automorphisms, it has a densely defined generator which is denoted as g_λ. The C*-algebra $\mathcal{F}$ with these *-automorphisms is called a field algebra [3, 24].

B. Decomposition of the system and initial local equilibrium states

We consider the situation where the system $\mathcal{S}$ can be decomposed into M independent infinitely extended subsystems $\mathcal{R}_j$ ($j = 1, \cdots M$), which play a role of reservoirs, and a finite-degree-of-freedom subsystem $\mathcal{S}_0$ interacting with all the others. More precisely, the algebra $\mathcal{F}$ is a tensor product of M infinite dimensional subalgebras $\mathcal{F}_j$ ($j = 1, \cdots M$), corresponding to $\mathcal{R}_j$, and a finite dimensional subalgebra $\mathcal{F}_S$, corresponding to $\mathcal{S}_0$:

$$\mathcal{F} = \mathcal{F}_S \otimes \mathcal{F}_1 \otimes \cdots \otimes \mathcal{F}_M \ , \tag{1}$$

such that the following conditions are satisfied:

(S1) There exists a gauge-invariant time evolution group τ_t^V ($t \in \mathbf{R}$) which is a perturbation of τ_t by a selfadjoint element $-V \in \mathcal{A} \cap D(\delta)$ and which is a product of strongly continuous groups $\tilde{\tau}_t^{(j)}$ ($j = 1, \cdots M$) independently acting on subalgebras $\mathcal{F}_j$ ($j = 1, \cdots M$)

$$\tau_t^V = \tilde{\tau}_t^{(1)} \cdots \tilde{\tau}_t^{(M)} . \tag{2}$$

Namely, $\tilde{\tau}_t^{(j)}$'s commute with each other and leave the elements of other subalgebras invariant:[30]

$$\tilde{\tau}_t^{(j)}(A) = A \qquad (^\forall A \in \mathcal{F}_k, \ k \neq j, \ k, j = 1, \cdots M) \tag{3}$$

$$\tilde{\tau}_t^{(j)} \tilde{\tau}_s^{(k)} = \tilde{\tau}_s^{(k)} \tilde{\tau}_t^{(j)} \quad (t, s \in \mathbf{R}, \ k \neq j, \ k, j = 1, \cdots M) . \tag{4}$$

(S2) The gauge *-automorphism $\alpha_{\vec{\varphi}}$ is a product of strongly continuous groups $\tilde{\alpha}_{\vec{\varphi}}^{(j)}$ ($j = 1, \cdots M$) and $\tilde{\alpha}_{\vec{\varphi}}^S$:

$$\alpha_{\vec{\varphi}} = \tilde{\alpha}_{\vec{\varphi}}^S \tilde{\alpha}_{\vec{\varphi}}^{(1)} \cdots \tilde{\alpha}_{\vec{\varphi}}^{(M)} , \tag{5}$$

where $\tilde{\alpha}_{\vec{\varphi}}^{(j)}$ and $\tilde{\alpha}_{\vec{\varphi}}^S$ independently act on subalgebras $\mathcal{F}_j$ ($j = 1, \cdots M$) and $\mathcal{F}_S$

$$\tilde{\alpha}_{\vec{\varphi}}^{(j)}(A) = A \qquad (^\forall A \in \mathcal{F}_k, \ k \neq j, \ k, j = S, 1, \cdots M) \tag{6}$$

$$\tilde{\alpha}_{\vec{\varphi}_1}^{(j)} \tilde{\alpha}_{\vec{\varphi}_2}^{(k)} = \tilde{\alpha}_{\vec{\varphi}_2}^{(k)} \tilde{\alpha}_{\vec{\varphi}_1}^{(j)} \quad (\vec{\varphi}_1, \vec{\varphi}_2 \in \mathbf{R}^L, \ k \neq j, \ k, j = S, 1, \cdots M) \tag{7}$$

And the groups $\tilde{\tau}_t^{(j)}$ and $\tilde{\alpha}_{\vec{\varphi}}^{(k)}$ commute with each other:

$$\tilde{\tau}_t^{(j)} \tilde{\alpha}_{\vec{\varphi}}^{(k)} = \tilde{\alpha}_{\vec{\varphi}}^{(k)} \tilde{\tau}_t^{(j)} \quad (j, k = 1, \cdots, M; \ t \in \mathbf{R}, \ \vec{\varphi} \in \mathbf{R}^L) .$$

States are introduced by listing expectation values. Namely, each state is identified with a linear map ω from $A \in \mathcal{F}$ to an expectation value $\omega(A)$. The positivity condition $\omega(A^*A) \geq 0$ and normalization condition $\omega(\mathbf{1}) = 1$ (with $\mathbf{1} \in \mathcal{F}$ the unity) are required. As in the previous works[4, 8, 10, 11, 15, 16, 17], we are interested in the evolution of initial states where M infinitely extended subsystems (reservoirs) are in equilibrium with different temperatures and different chemical potentials and the finite subsystem is in an arbitrary non-singular state. As discussed in [16, 17], such states are specified as a KMS state:

(S3) Let σ_x^ω ($x \in \mathbf{R}$) be a strongly continuous group defined by

$$\sigma_x^\omega(A) = \prod_{j=1}^M \tilde{\tau}_{-\beta_j x}^{(j)} \tilde{\alpha}_{\beta_j \vec{\mu}_j x}^{(j)} \left(e^{iD_S x} A e^{-iD_S x} \right) , \quad (A \in \mathcal{F}) \tag{8}$$

where β_j and $\vec{\mu}_j = (\mu_j^{(1)}, \cdots \mu_j^{(L)})$ are, respectively, the inverse temperature and the set of chemical potentials of the jth reservoir. The operator D_S ($\in \mathcal{F}_S \cap \mathcal{A}$) is selfadjoint. Then an initial state ω, which we are interested in, is a KMS state with temperature -1 with respect to σ_x^ω. Namely, ω is a state such that, for any pair $A, B \in \mathcal{F}$, there exists a function $F_{A,B}(x)$ of x-analytic in the stripe $\{x \in \mathbf{C}; 0 > \mathrm{Im} x > -1\}$ and satisfies the KMS boundary condition:

$$F_{A,B}(x) = \omega(A \sigma_x^\omega(B)) \qquad F_{A,B}(x - i) = \omega(\sigma_x^\omega(B) A) \quad (x \in \mathbf{R}) . \tag{9}$$

Hereafter, we assume that the domains of the generators $\tilde{\delta}_j$ and $\tilde{g}_\lambda^{(j)}$, respectively, of $\tilde{\tau}_t^{(j)}$ and $\tilde{\alpha}_{s\vec{e}_\lambda}^{(j)}$ ($t, s \in \mathbf{R}$) contain the domain $D(\delta)$ of the generator δ of τ_t:

(S4) $D(\delta) \subset D(\tilde{\delta}_j)$ $D(\delta) \subset D(\tilde{g}_\lambda^{(j)})$ ($^\forall j = 0, 1, \cdots M$, $^\forall \lambda = 1, \cdots L$).

Then, the condition (S1) implies that the domain of the generator δ^V of τ_t^V is identical with $D(\delta)$: $D(\delta^V) = D(\delta)$ and

$$\delta(A) = \delta^V(A) + i[V, A] \quad (A \in D(\delta)) \tag{10}$$

$$\delta^V(A) = \sum_{j=1}^N \tilde{\delta}_j(A) \qquad (A \in D(\delta)) \tag{11}$$

Moreover, the domain of the generator $\hat{\delta}_\omega$ of σ_x^ω also includes $D(\delta)$ and

$$\hat{\delta}_\omega(A) = -\sum_{j=1}^N \left\{ \beta_j \left(\tilde{\delta}_j(A) - \mu_\lambda^{(j)} \tilde{g}_\lambda^{(j)}(A) \right) \right\} + i[D_S, A] . \quad (A \in D(\delta)) \tag{12}$$

Note that a decomposition without the finite subsystem is possible as well.

C. L^1-asymptotic abelian property

In thermodynamics, environments are assumed to stay in equilibrium under arbitrary processes and their details are considered to be unimportant. Hence, thermodynamic environments would be well-modelled by systems with appropriate ergodicity. As one of such an example, we consider systems satisfying the L^1-asymptotic abelian property.

The time evolution τ_t is said to satisfy the $L^1(\mathcal{G})$-asymptotic abelian property if there exists a norm dense *-subalgebra $\mathcal{G}$ such that

$$\int_{-\infty}^{+\infty} dt \|[A, \tau_t(B)]\| \equiv \int_{-\infty}^{+\infty} dt \|A\tau_t(B) - \tau_t(B)A\| < +\infty \quad (A \in \mathcal{G}, \ B \in \mathcal{G} \cap \mathcal{F}_+)$$

$$\int_{-\infty}^{+\infty} dt \|[A, \tau_t(B)]_+\| \equiv \int_{-\infty}^{+\infty} dt \|A\tau_t(B) + \tau_t(B)A\| < +\infty \quad (A, B \in \mathcal{G} \cap \mathcal{F}_-)$$

where $\mathcal{F}_\pm$ are even/odd subalgebras. This property implies rapid decay of correlations and is satisfied by free fermions in $\mathbf{R}^d$ ($d \geq 1$) (Example 5.4.9 of Ref. [3]). Note that, if a system admits bound states, it does not satisfy the L^1-asymptotic abelian condition as there exist bounded constants of motion, i.e., observables C satisfying $\tau_t(C) = C$.

III.   NESS AND MACLENNAN-ZUBAREV ENSEMBLES

The state ω_t at time t starting from the initial state ω is given by

$$\omega_t(A) = \omega \left(\tau_t(A) \right) , \quad (^\forall A \in \mathcal{F}) \tag{13}$$

and, under the setting (S1)-(S4), nonequilibrium steady states are expected to be obtained as its weak limits for $t \to \pm\infty$. As shown by Ruelle[9], it is indeed the case. Namely, under the setting

(S1)-(S4), if the time evolution τ_t is $L^1(\mathcal{G})$-asymptotic abelian and the perturbation V is an element of $\mathcal{G}$, the limits

$$\lim_{t\to\pm\infty} \omega\left(\tau_t(A)\right) = \omega(\gamma_\pm(A)) \equiv \omega_\pm(A)$$

exist for all $A \in \mathcal{F}$ and define nonequilibrium steady states, where $\gamma_\pm$ are Møller morphisms defined by

$$\lim_{t\to\pm\infty} \left\|\tau_t^{V^{-1}}\tau_t(A) - \gamma_\pm(A)\right\| = 0 \quad (^\forall A \in \mathcal{F}) \ .$$

Also Ruelle showed the independence of the limits on the way of separating the whole system into reservoirs and a small system[9]. See also Refs. [11, 12]. In this section, as announced in Ref. [20], under a stronger condition, these steady states $\omega_\pm$ are shown to be inerpreted as MacLennan-Zubarev ensembles[21, 22]. Indeed, one has:

Proposition 1: KMS characterization of evolving states

Under the setting (S1)-(S4), the state ω_t at time t is a KMS state at temperature -1 with respect to the strongly continuous group of *-automorphisms

$$\sigma_x^{\omega_t} \equiv \gamma_t^{-1}\sigma_x^\omega\gamma_t \ , \tag{14}$$

where $\gamma_t = \tau_t^{V^{-1}}\tau_t$, and its generator is given by

$$\hat{\delta}_\omega^{(t)}(A) = \hat{\delta}_\omega(A) + i\int_{-t}^0 ds \left[\tau_s\left(\hat{\delta}_\omega(V)\right), A\right] \ , \quad (^\forall A \in D(\hat{\delta}_\omega^{(t)}) = D(\hat{\delta}_\omega)) \ . \tag{15}$$

Proposition 2: KMS characterization of steady states

Under the setting (S1)-(S4), if the time evolution *-automorphism τ_t is $L^1(\mathcal{G})$-asymptotically abelian, $V \in \mathcal{G}$ and the Møller morphisims $\gamma_\pm$ are invertible, the steady states $\omega_\pm$ are KMS states at temperature -1 with respect to the strongly continuous group of *-automorphisms

$$\sigma_x^{\omega_\pm} \equiv \gamma_\pm^{-1}\sigma_x^\omega\gamma_\pm \ . \tag{16}$$

Furthermore, if $\hat{\delta}_\omega(V) \in \mathcal{G}$, its generator $\hat{\delta}_\omega^\pm$ satisfies

$$\hat{\delta}_\omega^\pm(A) = \hat{\delta}_\omega(A) + i\int_{\mp\infty}^0 ds \left[\tau_s\left(\hat{\delta}_\omega(V)\right), A\right] \ , \quad (^\forall A \in D(\hat{\delta}_\omega) \cap \mathcal{G}) \ . \tag{17}$$

NB 3 For finite systems, the KMS state ω with respect to the *-automorphism σ_x^ω corresponds to the density matrix

$$\rho_\omega = \frac{1}{Z}\exp\left\{-\sum_{j=1}^N \beta_j\left(H_j - \sum_{\lambda=1}^L \mu_\lambda^{(j)} N_j^{(\lambda)}\right)\right\} \ ,$$

where Z is the normalization constant, β_j, H_j, $\mu_\lambda^{(j)}$ and $N_j^{(\lambda)}$ are, respectively, the local temperature, local energy, local chemical potential and local number operator of the jth reservoir. Then, as a result of the Liouville-von Neumann eqation, the density matrix at time t is given by

$$\tau_t^{-1}(\rho_\omega) = \frac{1}{Z}\exp\left\{-\sum_{j=1}^M \beta_j\left(\tau_{-t}(H_j) - \sum_{\lambda=1}^L \mu_\lambda^{(j)}\tau_{-t}(N_j^{(\lambda)})\right)\right\}$$

$$= \frac{1}{Z} \exp\Big\{-\sum_{j=1}^{M} \beta_j \Big[H_j - \sum_{\lambda=1}^{L} \mu_\lambda^{(j)} N_j^{(\lambda)} - \int_{-t}^{0} ds\tau_s(J_j^q)\Big]\Big\} \, ,$$

where $J_j^q \equiv \frac{d}{dt}\tau_t\left(H_j\right) - \sum_{\lambda=1}^{L} \mu_\lambda^{(j)} \frac{d}{dt}\tau_t(N_j^{(\lambda)})\Big|_{t=0} = -i[H_j, V] + \sum_{\lambda=1}^{L} \mu_\lambda^{(j)} i[N_j^{(\lambda)}, V]$ is non-systematic energy flow, or heat flow, to the jth reservoir and we have used

$$\tau_{-t}(H_j) = H_j - \int_{-t}^{0} ds\frac{d}{ds}\tau_s(H_j) \, , \quad \tau_{-t}(N_j^{(\lambda)}) = N_j^{(\lambda)} - \int_{-t}^{0} ds\frac{d}{ds}\tau_s(N_j^{(\lambda)}) \, .$$

On the other hand, as $\tilde{\delta}_j(V) = i[H_j, V]$ and $\tilde{g}_\lambda^{(j)} = i[N_j^{(\lambda)}, V]$, one has

$$\sum_{j=1}^{M} \beta_j \tau_s(J_j^q) = -\tau_s\Big(\sum_{j=1}^{M} \beta_j\{\tilde{\delta}_j(V) - \sum_{\lambda=1}^{L} \mu_\lambda^{(j)} \tilde{g}_\lambda^{(j)}(V)\}\Big) = -\tau_s\left(\hat{\delta}_\omega(V)\right)$$

and, thus,

$$\tau_t^{-1}(\rho_\omega) = \frac{1}{Z} \exp\Big\{-\sum_{j=1}^{M} \beta_j \Big[H_j - \sum_{\lambda=1}^{L} \mu_\lambda^{(j)} N_j^{(\lambda)} + \int_{-t}^{0} ds\tau_s\left(\hat{\delta}_\omega(V)\right)\Big]\Big\} \, ,$$

which is a KMS state generated by $\hat{\delta}_\omega(A) + i\int_{-t}^{0} ds[\tau_s\left(\hat{\delta}_\omega(V)\right), A]$. This observation is nothing but the finite dimensional version of Proposition 1.

NB 4 For infinite systems, an interesting case is that where the right-hand side of (17) generates $\sigma_x^{\omega\pm}$. Then, if the integral

$$\widetilde{V}_\pm \equiv \int_{\mp\infty}^{0} ds\tau_s\left(\hat{\delta}_\omega(V)\right) \tag{18}$$

would converge, $\omega_\pm$ would be perturbed KMS states of the initial state ω by self-adjoint operators $\widetilde{V}_\pm$. Moreover, NB 3 suggests that the corresponding density matrices would be

$$\rho_\pm = \frac{1}{Z} \exp\Big\{-\sum_{j=1}^{N} \beta_j \Big[H_j - \sum_{\lambda=1}^{L} \mu_\lambda^{(j)} N_j^{(\lambda)} - \int_{\mp\infty}^{0} ds\tau_s(J_j^q)\Big]\Big\} \, . \tag{19}$$

Note that such ensembles for steady states were introduced by MacLennan[21] and Zubarev[22].

However, if the steady states carry nonvanishing entropy production, the integral $\widetilde{V}_\pm$ does not converge since the steady-state average of its integrand is nothing but the nonvanishing enropy production rate at the steady states:

$$\lim_{s\to\pm\infty} \omega\left(\tau_s\left(\hat{\delta}_\omega(V)\right)\right) = \omega_\pm\Big(\sum_{j=1}^{M} \beta_j J_j^q\Big) \neq 0 \, .$$

This observation is consistent with the results by Jakšić and Pillet[17] who showed that, if the steady-state entropy production is nonvanishing, the steady state is 'singular' with respect to the initial local equilibrium state. Thus, the original proposal (19) by MacLennan and Zubarev cannot be justified. Rather, the KMS states with respect to $\sigma_x^{\omega\pm}$ generated by (17) should be regarded as a precise definition of the MacLennan-Zubarev ensembles.

Proof of Proposition 1

As easily seen from (S1) and (S4), the initial state ω is invariant under τ_t^V. And one has $\omega_t(A) = \omega\left(\tau_t(A)\right) = \omega\left(\gamma_t(A)\right)$ ($^\forall A \in \mathcal{F}$). Since γ_t is a *-automorphism and ω is a KMS state with respect to σ_x^ω at temperature -1, ω_t is a KMS state with respect to $\gamma_t^{-1}\sigma_x^\omega\gamma_t$ at temperature -1 (cf. Prop. 5.3.33 of Ref. [3]).

Now we consider the generator. In terms of the one-parameter family Y_t of unitary elements defined as a norm convergent series:

$$Y_t = \mathbf{1} + \sum_{n=1}^{+\infty} i^n \int_0^t dt_1 \int_0^{t_1} dt_2 \cdots \int_0^{t_{n-1}} dt_n \tau_{-t_n}(V) \cdots \tau_{-t_2}(V)\tau_{-t_1}(V) , \tag{20}$$

one has $\gamma_t(A) = Y_t A Y_t^*$. Thus, $\gamma_t(A)$ ($^\forall A \in \mathcal{F}$) is differentiable with respect to t and $\dfrac{d}{dt}\gamma_t(A) = i\gamma_t\left([\tau_{-t}(V), A]\right)$. This leads to

$$\frac{d\{\gamma_t^{-1}\sigma_x^\omega\gamma_t(A)\}}{dt} = i[\gamma_t^{-1}\sigma_x^\omega\gamma_t\tau_{-t}(V) - \tau_{-t}(V), \gamma_t^{-1}\sigma_x^\omega\gamma_t(A)] = i\left[\tau_{-t}\left(\sigma_x^\omega(V) - V\right), \gamma_t^{-1}\sigma_x^\omega\gamma_t(A)\right]$$

or

$$\gamma_t^{-1}\sigma_x^\omega\gamma_t(A) = \sigma_x^\omega(A) + i \int_0^t ds \left[\tau_{-s}\left(\sigma_x^\omega(V) - V\right), \gamma_s^{-1}\sigma_x^\omega\gamma_s(A)\right] , \tag{21}$$

where $\gamma_t\tau_{-t} = \tau_{-t}^V$, $\tau_{-t}^V\sigma_x^\omega = \sigma_x^\omega\tau_{-t}^V$ and $\gamma_t^{-1}\tau_{-t}^V = \tau_{-t}$ have been used. Since $V \in D(\delta) \subset D(\hat{\delta}_\omega)$, one has the following relation for any $A \in D(\hat{\delta}_\omega)$:

$$\begin{aligned}
&\left.\frac{d}{dx}\gamma_t^{-1}\sigma_x^\omega\gamma_t(A)\right|_{x=0} \\
&= \lim_{x\to 0}\left\{\frac{\sigma_x^\omega(A) - A}{x} + i \int_0^t ds\left[\tau_{-s}\left(\frac{\sigma_x^\omega(V) - V}{x}\right), \gamma_s^{-1}\sigma_x^\omega\gamma_s(A)\right]\right\} \\
&= \hat{\delta}_\omega(A) + i \int_{-t}^0 ds\left[\tau_s\left(\hat{\delta}_\omega(V)\right), A\right] .
\end{aligned} \tag{22}$$

This suggests that $\widetilde{\delta}_{\omega_t}$ defined by

$$\widetilde{\delta}_{\omega_t}(A) \equiv \hat{\delta}_\omega(A) + i \int_{-t}^0 ds\left[\tau_s\left(\hat{\delta}_\omega(V)\right), A\right]$$

is the generator $\hat{\delta}_\omega^{(t)}$ of $\gamma_t^{-1}\sigma_x^\omega\gamma_t$. As we see, it is the case. First, we note that, since $\int_{-t}^0 ds\tau_s\left(\hat{\delta}_\omega(V)\right)$ is selfadjoint, $\widetilde{\delta}_{\omega_t}$ generates a strongly continuous group (cf. Theorem 4.1 of Ref. [3]). Then as a result of the theory of semigroups (Prop. 3.1.6 of Ref. [3]), its resolvent $(\mu 1 + \widetilde{\delta}_{\omega_t})^{-1}$ ($\mu \in \mathbf{R}\backslash\{0\}$) is bounded, where 1 stands for the identity operator on $\mathcal{F}$. For any $B \in \mathcal{F}$, let $A = (\mu 1 + \widetilde{\delta}_{\omega_t})^{-1}B$, then $A \in D(\hat{\delta}_\omega)$ and (22) leads to

$$(\mu 1 + \hat{\delta}_\omega^{(t)})A = \mu A + \left.\frac{d}{dx}\gamma_t^{-1}\sigma_x^\omega\gamma_t(A)\right|_{x=0} = B .$$

On the other hand, as a generator of strongly continuous group $\gamma_t^{-1}\sigma_x^\omega\gamma_t$, the resolvent $(\mu 1 + \hat{\delta}_\omega^{(t)})^{-1}$ of $\hat{\delta}_\omega^{(t)}$ is again bounded. Therefore, one finally has

$$(\mu 1 + \hat{\delta}_\omega^{(t)})^{-1}B = A = (\mu 1 + \widetilde{\delta}_{\omega_t})^{-1}B$$

for all $B \in \mathcal{F}$ and $\hat{\delta}_\omega^{(t)} = \widetilde{\delta}_{\omega_t}$. Moreover, as $D(\widetilde{\delta}_{\omega_t}) = D(\hat{\delta}_\omega)$, $D(\hat{\delta}_\omega^{(t)}) = D(\hat{\delta}_\omega)$.

Proof of Proposition 2

Remind that $\omega_\pm(A) = \omega\left(\gamma_\pm(A)\right)$ ($^\forall A \in \mathcal{F}$). When the Møller morphisms $\gamma_\pm$ are invertible, they are *-automorphisms. Thus, since ω is a KMS state with respect to σ_x^ω at temperature -1, $\omega_\pm$ is a KMS state with respect to $\gamma_\pm^{-1}\sigma_x^\omega\gamma_\pm$ at temperature -1 (cf. Prop. 5.3.33 of Ref. [3]).

Now we show the second half. Since *-automorphisms are norm preserving (cf. e.g. Corollary 2.3.4 of Ref. [3]), one has

$$\|\gamma_{\pm n}^{-1}\sigma_x^\omega\gamma_{\pm n}(A) - \gamma_\pm^{-1}\sigma_x^\omega\gamma_\pm(A)\| \le \|\gamma_{\pm n}(A) - \gamma_\pm(A)\| + F(x,n) \ , \tag{23}$$

where $\gamma_{\pm n} = {\tau_{\pm n}^V}^{-1}\tau_{\pm n}$ and

$$F(x,n) = \left\|\gamma_{\pm n}\left(\gamma_\pm^{-1}\sigma_x^\omega\gamma_\pm(A)\right) - \gamma_\pm\left(\gamma_\pm^{-1}\sigma_x^\omega\gamma_\pm(A)\right)\right\| \ .$$

As the Møller morphism is a strong operator limit of $\gamma_{\pm n}$,

$$\lim_{n\to+\infty} \|\gamma_{\pm n}(A) - \gamma_\pm(A)\| = 0, \qquad (^\forall A \in \mathcal{F}) \tag{24}$$

$$\lim_{n\to+\infty} F(x,n) = 0 \ . \qquad \text{(for each } x) \tag{25}$$

Next, as a result of

$$\begin{aligned}|F(x,n) - F(x',n)| &\le \|\gamma_{\pm n}\gamma_\pm^{-1}\left\{\sigma_x^\omega\gamma_\pm(A) - \sigma_{x'}^\omega\gamma_\pm(A)\right\}\| \\ &= \|\sigma_x^\omega\gamma_\pm(A) - \sigma_{x'}^\omega\gamma_\pm(A)\| \ ,\end{aligned}$$

the function $x \to F(x,n)$ is continuous uniformly with respect to n. Hence, the limit (25) is uniform with respect to x on any finite interval in $\mathbf{R}$. In short,

$$\lim_{n\to+\infty} \|\gamma_{\pm n}^{-1}\sigma_x^\omega\gamma_{\pm n}(A) - \gamma_\pm^{-1}\sigma_x^\omega\gamma_\pm(A)\| = 0 \ , \tag{26}$$

where the convergence is uniform with respect to x on any finite interval in $\mathbf{R}$. Hence, as a result of Theorem 3.1.28 of Ref. [3] , the generator $\hat{\delta}_\omega^\pm$ of $\gamma_\pm^{-1}\sigma_x^\omega\gamma_\pm$ is the graph limit of the generators $\hat{\delta}_\omega^{(\pm n)}$ of $\gamma_{\pm n}^{-1}\sigma_x^\omega\gamma_{\pm n}$. Namely, if a sequence $\{A_n\}_{n=1}^{+\infty}$ with $A_n \in D(\hat{\delta}_\omega^{(\pm n)})$ satisfies $A_n \to A$ and $\hat{\delta}_\omega^{(\pm n)}(A_n) \to B$ $(n \to +\infty)$, then one has $B = \hat{\delta}_\omega^\pm(A)$.

For each $A \in D(\hat{\delta}_\omega) \cap \mathcal{G}$, let $A_n = A$, then obviously $\lim_{n\to+\infty} A_n = A$, $A_n \in D(\hat{\delta}_\omega^{(\pm n)})$ because of Proposition 1. And, as $\hat{\delta}_\omega(V) \in \mathcal{G}$ is assumed, the $L^1(\mathcal{G})$-asymptotic abelian property of τ_t implies

$$\lim_{n\to+\infty} \hat{\delta}_\omega^{(\pm n)}(A_n) = \hat{\delta}_\omega(A) + i\int_{\mp\infty}^0 ds\left[\tau_s\left(\hat{\delta}_\omega(V)\right), A\right] \ .$$

Therefore, one has the desired result

$$\hat{\delta}_\omega^\pm(A) = \hat{\delta}_\omega(A) + i\int_{\mp\infty}^0 ds\left[\tau_s\left(\hat{\delta}_\omega(V)\right), A\right] \ .$$

IV. QUASISTATIC PROCESS AND CLAUSIUS EQUALITY

In this section, we consider the case where $\mathcal{F}_S$ is the algebra of bounded operators on a finite dimensional Hilbert space and the small system couples with a single reservoir, namely the case of $M = 1$. Then, the total system is described by the tensor product $\mathcal{F}_S \otimes \mathcal{F}_1$ and the 'decoupled' evolution τ_t^V acts only on the reservoir algebra: $\tau_t^V \equiv \tilde{\tau}_t^{(1)}$. Since the system is finite dimensional,

the generator of the gauge transformation $\tilde{\alpha}^{(S)}_{s\vec{e}_\lambda}$ is a commutator with a self-adjoint element $N_\lambda \otimes \mathbf{1}_1$ where $N_\lambda \in \mathcal{F}_S$ is the number operator of the λth particles and $\mathbf{1}_1$ is the unit of $\mathcal{F}_1$. The system-reservoir interaction is assumed to be proportional to a coupling constant κ: κV where $V \in D(\delta)$ is self-adjoint and gauge-invariant: $\alpha_{\vec{\varphi}}(V) = V$. Note that the gauge invariance of V leads to $\tilde{g}^{(1)}_\lambda(V) + i[N_\lambda \otimes \mathbf{1}_1, V] = 0$ $(\lambda = 1, \cdots L)$.

We are interested in the response of the whole system under a time-dependent perturbation: $W(t) \otimes \mathbf{1}_1$ where $W(t) \in \mathcal{F}_S$ is twice continuously differentiable in norm, $W(t) = W_0$ for $t \le 0$ and $[N_\lambda \otimes \mathbf{1}_1, W(t) \otimes \mathbf{1}_1] = 0$. Initially, the whole system is prepared to be an equilibrium state ω of the inverse temperature β and the chemical potentials μ_λ, namely, a KMS state at β with respect to $\hat{\sigma}_s \equiv \alpha^{(i)}_s \alpha_{-s\vec{\mu}}$ where $\alpha^{(i)}_t$ is defined by

$$\frac{d\alpha^{(i)}_t(A)}{dt} = \alpha^{(i)}_t \left(\tilde{\delta}_1(A) + i[W_0 \otimes \mathbf{1}_1 + \kappa V, A] \right) , \quad \alpha^{(i)}_t(A)|_{t=0} = A , \quad (\forall A \in D(\tilde{\delta}_1)) .$$

The time evolution τ^W_t is given by the solution of

$$\frac{d\tau^W_t(A)}{dt} = \tau^W_t \left(\tilde{\delta}_1(A) + i[\kappa V + W(t) \otimes \mathbf{1}_1, A] \right) , \quad \tau^W_t(A)|_{t=0} = A , \quad (A \in D(\tilde{\delta}_1)) ,$$

and the state at time t by $\omega_t(A) \equiv \omega \left(\tau^W_t(A) \right)$ $(\forall A \in \mathcal{F})$. Now, we define the the system-energy increase Z_T induced by the mass flow, the work W_T done on the system and the heat Q_T absorbed by the system during the time interval T as follows:

$$
\begin{aligned}
Z_T &= \sum_{\lambda=1}^L \mu_\lambda \left\{ \omega_T(N_\lambda \otimes \mathbf{1}_1) - \omega(N_\lambda \otimes \mathbf{1}_1) \right\} \\
W_T &= \int_0^T dt\, \omega_t \left(\tfrac{d}{dt} W(t) \otimes \mathbf{1}_1 \right) \\
Q_T &= \left\{ \omega_T(W(T) \otimes \mathbf{1}_1 + \kappa V) - \omega(W_0 \otimes \mathbf{1}_1 + \kappa V) \right\} - W_T - Z_T
\end{aligned}
$$

Then, one has the following results:

Proposition 5: Stepwise perturbation
Suppose $W(t) = W_f$ $(t \ge t_0 > 0)$. Let $\alpha^{(f)}_t$ be the evolution defined by

$$\frac{d\alpha^{(f)}_t(A)}{dt} = \alpha^{(f)}_t \left(\tilde{\delta}_1(A) + i[W_f \otimes \mathbf{1}_1 + \kappa V, A] \right) , \quad \alpha^{(f)}_t(A)|_{t=0} = A , \quad (A \in D(\tilde{\delta}_1)) ,$$

ω_f be a KMS state with respect to $\hat{\sigma}^{(f)}_s \equiv \alpha^{(f)}_s \alpha_{-s\vec{\mu}}$ at β, and $(\mathcal{H}_f, \pi_f, \Omega_f)$ be its GNS representation[31]. Then, if the initial state ω is the unique KMS state and a self-adjoint operator L_f on $\mathcal{H}_f$ defined by $e^{iL_f t}\pi_f(A)\Omega_f \equiv \pi_f(\alpha^{(f)}_t(A))\Omega_f$ $(A \in \mathcal{F})$, which will be called the Liouvillian of $\alpha^{(f)}_t$, has a simple eigenvalue at zero and the absolute continuous spectrum, one has

$$\lim_{\kappa \to 0} \lim_{T \to +\infty} \beta Q_T = S(\rho_f) - S(\rho_i) - S(\rho_f | \rho_{t_0})$$

where $S(\rho_\nu) = -\mathrm{Tr}\{\rho_\nu \ln \rho_\nu\}$ $(\nu = i, f)$ is the von Neumann entropy of the density matrix ρ_ν, $S(\rho_f | \rho_{t_0}) = \mathrm{Tr}\{\rho_{t_0}(\ln \rho_{t_0} - \ln \rho_f)\} \ge 0$ is the relative entropy between density matrices ρ_f and ρ_{t_0}, and $\rho_i = \exp(-\beta(W_0 - \sum_\lambda \mu_\lambda N_\lambda))/\Xi_i$, $\rho_{t_0} = u^*_{t_0}\rho_i u_{t_0}$, $\rho_f = \exp(-\beta(W_f - \sum_\lambda \mu_\lambda N_\lambda))/\Xi_f$, with Ξ_i, Ξ_f the grand partition functions. The unitary element $u_t \in \mathcal{F}_S$ is the solution of $\frac{d}{dt}u_t = iu_t W(t)$, $u_t|_{t=0} = \mathbf{1}_S$.

Proposition 6: Staircase perturbation

Suppose that $T = \sum_{j=1}^{N} T_j$ and the interaction $W(t)$ has a staircase form: $W(t) = W_0 + (j-1+\varphi(t-\tilde{T}_{j-1}))(W_f-W_0)/N$ for $\tilde{T}_{j-1} \leq t \leq \tilde{T}_j$ where $\tilde{T}_j = \sum_{k=1}^{j} T_k, (\tilde{T}_0 \equiv 0)$ and $\varphi(t)$ is a twice continuously differentiable real-valued function with $\varphi(0) = 0$, $\varphi(t) = 1$ for $t \geq t_0$. Define $W_j \equiv W_0 + j(W_f - W_0)/N$ and introduce the group $\alpha_t^{(j)}$ by

$$\frac{d\alpha_t^{(j)}(A)}{dt} = \alpha_t^{(j)}\left(\tilde{\delta}_1(A) + i[W_j \otimes \mathbf{1}_1 + \kappa V, A]\right) , \quad \alpha_t^{(j)}(A)|_{t=0} = A , \quad (^\forall A \in D(\tilde{\delta}_1)) .$$

Let ω_j be a KMS state with respect to $\hat{\sigma}_s^{(j)} \equiv \alpha_s^{(j)}\alpha_{-s\vec{\mu}}$ at β, and $(\mathcal{H}_j, \pi_j, \Omega_j)$ be its GNS representation. Then, if the initial state ω is the unique KMS state and the Liouvillian L_j defined by $e^{iL_jt}\pi_j(A)\Omega_j \equiv \pi_j(\alpha_t^{(j)}(A))\Omega_j$ $(A \in \mathcal{F})$ has a simple eigenvalue at zero and the absolute continuous spectrum, one has

$$\lim_{\kappa \to 0} \lim_{T_1 \to +\infty} \cdots \lim_{T_N \to +\infty} \beta Q_T = S(\rho_f) - S(\rho_i) + \mathrm{O}\left(\frac{1}{N}\right)$$

NB 7: Because of the return-to-equilibrium property, the final state (and every intermediate state in Proposition 6) of the whole system is an equlibrium state. The limit of $\kappa \to 0$ implies that the coupling between the system and the reservoir is negligibly small. Thus, the processes treated in Proposition 5 and Proposition 6 precisely correspond to those in the classical thermodynamics [26]. The Clausius inequality follows from Proposition 5: $\lim_{\kappa \to 0} \lim_{T \to +\infty} \beta Q_T \leq S(\rho_f) - S(\rho_i)$ because of the positivity of the relative entropy. On the other hand, Proposition 6 implies that, if the whole system changes very slowly ($N \gg 1$) so that the whole system is in equlibrium at every instant, the Clausius equality holds. Thus, the thermodynamic entropy is given by the von Neumann entropy, as expected, and the process described in Proposition 6 is nothing but a quasistatic process. Note that, as κV is responsible for the equlibration, the weak coupling limit $\kappa \to 0$ should be taken *after* the long term limits.

NB 8: The two propositions deal with the entropy change of a subsystem in contrast to the previous works [9, 11, 15, 16, 17, 18, 27], which have discussed the entropy production of the whole system. In this respect, the present work shares a common interest with that by Maes and Tasaki[28], who derived a relation equivalent to the Clausius inequality from dynamics for a class of finite classical systems. The difference between the present setting and that of Ref. [28] lies in the fact that we deal with an infinite-degree-of-freedom reservoir in order to have equilibrium states as the final state of Proposition 5 and as the intermediate states of Proposition 6.

NB 9: Fröhlich, Merkli, Schwarz, and Ueltschi[27] have discussed the Clausius equality for 'adiabatic' processes where the difference between the true and the reference states[32] restricted to a certain subsystem is small for all times. However, conditions on the dynamics which realize the 'adiabatic' processes are not given. Instead, Proposition 6 gives a concrete example of quasi-static processes, which is similar to the one studied for the classical stochastic systems by Sekimoto [29].

Proof of Proposition 5: The heat is rewritten and, then, the assertions are shown.

Expression of heat: Let Γ_t be solutions of $\frac{d}{dt}\Gamma_t = i\Gamma_t\tilde{\tau}_t^{(1)}(W(t) \otimes \mathbf{1}_1 + \kappa V)$, $\Gamma_t|_{t=0} = \mathbf{1}$, where $\mathbf{1} \equiv \mathbf{1}_S \otimes \mathbf{1}_1$, then $\tau_t^W(A) = \Gamma_t\tilde{\tau}_t^{(1)}(A)\Gamma_t^*$ and

$$-i\frac{d}{dt}\left((\tilde{\delta}_1 - \sum_{\lambda=1}^{L}\mu_\lambda\tilde{g}_\lambda^{(1)})(\Gamma_t)\Gamma_t^*\right) = (\tilde{\delta}_1 - \sum_{\lambda=1}^{L}\mu_\lambda\tilde{g}_\lambda^{(1)})(\Gamma_t\tilde{\tau}_t^{(1)}(W(t) \otimes \mathbf{1}_1 + \kappa V))\Gamma_t^*$$

$$-(\tilde{\delta}_1 - \sum_{\lambda=1}^{L}\mu_\lambda\tilde{g}_\lambda^{(1)})(\Gamma_t)\tilde{\tau}_t^{(1)}(W(t) \otimes \mathbf{1}_1 + \kappa V)\Gamma_t^* = \tau_t^W\left((\tilde{\delta}_1 - \sum_{\lambda=1}^{L}\mu_\lambda\tilde{g}_\lambda^{(1)})(\kappa V)\right) \quad (27)$$

where we have used $(\tilde{\delta}_1 - \sum_{\lambda=1}^{L} \mu_\lambda \tilde{g}_\lambda^{(1)})(AB) = (\tilde{\delta}_1 - \sum_{\lambda=1}^{L} \mu_\lambda \tilde{g}_\lambda^{(1)})(A)B + A(\tilde{\delta}_1 - \sum_{\lambda=1}^{L} \mu_\lambda \tilde{g}_\lambda^{(1)})(B)$, $\tilde{\delta}_1(W(t) \otimes \mathbf{1}_1) = 0$ and $\tilde{g}_\lambda^{(1)}(W(t) \otimes \mathbf{1}_1) = 0$. Reminding

$$
\begin{aligned}
\frac{d}{dt} \tau_t^W (N_\lambda \otimes \mathbf{1}_1) &= \tau_t^W \left(\tilde{\delta}_1(N_\lambda \otimes \mathbf{1}_1) + i[W(t) \otimes \mathbf{1}_1 + \kappa V, N_\lambda \otimes \mathbf{1}_1] \right) \\
&= \tau_t^W \left(i[\kappa V, N_\lambda \otimes \mathbf{1}_1] \right) = \tau_t^W (\tilde{g}_\lambda^{(1)}(\kappa V)) ,
\end{aligned} \tag{28}
$$

$$
\begin{aligned}
\frac{d}{dt} \tau_t^W (W(t) \otimes \mathbf{1}_1 + \kappa V) &= \tau_t^W \left(\tilde{\delta}_1(W(t) \otimes \mathbf{1}_1 + \kappa V) + i[W(t) \otimes \mathbf{1}_1 + \kappa V, W(t) \otimes \mathbf{1}_1 + \kappa V] \right) \\
&\quad + \tau_t^W \left(\frac{dW(t)}{dt} \otimes \mathbf{1}_1 \right) = \tau_t^W \left(\tilde{\delta}_1(\kappa V) \right) + \tau_t^W \left(\frac{dW(t)}{dt} \otimes \mathbf{1}_1 \right) ,
\end{aligned} \tag{29}
$$

one obtains

$$
\begin{aligned}
&-i \frac{d}{dt} \left((\tilde{\delta}_1 - \sum_{\lambda=1}^{L} \mu_\lambda \tilde{g}_\lambda^{(1)})(\Gamma_t) \Gamma_t^* \right) \\
&= \frac{d\tau_t^W (W(t) \otimes \mathbf{1}_1 + \kappa V)}{dt} - \tau_t^W \left(\frac{dW(t)}{dt} \otimes \mathbf{1}_1 \right) - \sum_{\lambda=1}^{L} \mu_\lambda \frac{d\tau_t^W (N_\lambda \otimes \mathbf{1}_1)}{dt}
\end{aligned} \tag{30}
$$

and, thus,

$$
\begin{aligned}
Q_T &\equiv \omega(\tau_T^W (W(T) \otimes \mathbf{1}_1 + \kappa V)) - \omega(W(0) \otimes \mathbf{1}_1 + \kappa V) \\
&\quad - \int_0^T ds\, \omega \left(\tau_s^W \left(\frac{dW(s)}{ds} \otimes \mathbf{1}_1 \right) \right) - \sum_{\lambda=1}^{L} \mu_\lambda \left(\omega(\tau_T^W (N_\lambda \otimes \mathbf{1}_1)) - \omega(N_\lambda \otimes \mathbf{1}_1) \right) \\
&= -i\omega \left((\tilde{\delta}_1 - \sum_{\lambda=1}^{L} \mu_\lambda \tilde{g}_\lambda^{(1)})(\Gamma_T) \Gamma_T^* \right) .
\end{aligned} \tag{31}
$$

Proof of the assertions: Let $\Gamma_t^{(f)}$ be the solution of $\frac{d}{dt}\Gamma_t^{(f)} = i\Gamma_t^{(f)} \tilde{\tau}_t^{(1)}(W_f \otimes \mathbf{1}_1 + \kappa V)$, $\Gamma_t^{(f)}|_{t=0} = \mathbf{1}$, then, as in the derivation of (30), one obtains $\alpha_t^{(f)}(A) = \Gamma_t^{(f)} \tilde{\tau}_t^{(1)}(A) \Gamma_t^{(f)*}$,

$$
\begin{aligned}
&-i \frac{d}{dt} \left\{ (\tilde{\delta}_1 - \sum_{\lambda=1}^{L} \mu_\lambda \tilde{g}_\lambda^{(1)})(\Gamma_t^{(f)}) \Gamma_t^{(f)*} \right\} = \alpha_t^{(f)} \left((\tilde{\delta}_1 - \sum_{\lambda=1}^{L} \mu_\lambda \tilde{g}_\lambda^{(1)})(\kappa V) \right) \\
&= \frac{d}{dt} \alpha_t^{(f)} \left(W_f \otimes \mathbf{1}_1 + \kappa V - \sum_{\lambda=1}^{L} \mu_\lambda N_\lambda \otimes \mathbf{1}_1 \right)
\end{aligned}
$$

and, thus,

$$
-i(\tilde{\delta}_1 - \sum_{\lambda=1}^{L} \mu_\lambda \tilde{g}_\lambda^{(1)})(\Gamma_t^{(f)}) \Gamma_t^{(f)*} = \alpha_t^{(f)} \left((W_f - \sum_{\lambda=1}^{L} \mu_\lambda N_\lambda) \otimes \mathbf{1}_1 + \kappa V \right) - (W_f - \sum_{\lambda=1}^{L} \mu_\lambda N_\lambda) \otimes \mathbf{1}_1 - \kappa V .
$$

Then, because of $\Gamma_t = \Gamma_{t_0} \tilde{\tau}_{t_0}^{(1)} \left(\Gamma_{t-t_0}^{(f)} \right)$ for $t \geq t_0$, the heat flow to the reservoir is rewritten as

$$
\begin{aligned}
Q_T &= -i\omega \left(\Gamma_{t_0} \tilde{\tau}_{t_0}^{(1)} \left((\tilde{\delta}_1 - \sum_{\lambda=1}^{L} \mu_\lambda \tilde{g}_\lambda^{(1)})(\Gamma_{T-t_0}^{(f)}) \Gamma_{T-t_0}^{(f)*} \right) \Gamma_{t_0}^* \right) - i\omega \left((\tilde{\delta}_1 - \sum_{\lambda=1}^{L} \mu_\lambda \tilde{g}_\lambda^{(1)})(\Gamma_{t_0}) \Gamma_{t_0}^* \right) \\
&= \omega \left(\Gamma_{-t_0}^{(i)} \bar{\Gamma}_{t_0} \alpha_{T-t_0}^{(f)} ((W_f - \sum_{\lambda=1}^{L} \mu_\lambda N_\lambda) \otimes \mathbf{1}_1 + \kappa V) \bar{\Gamma}_{t_0}^* \Gamma_{-t_0}^{(i)*} \right) \\
&\quad - \omega \left(\Gamma_{t_0} \tilde{\tau}_{t_0}^{(1)} ((W_f - \sum_{\lambda=1}^{L} \mu_\lambda N_\lambda) \otimes \mathbf{1}_1) \Gamma_{t_0}^* \right) - \omega \left(\Gamma_{t_0} \tilde{\tau}_{t_0}^{(1)}(\kappa V) \Gamma_{t_0}^* \right)
\end{aligned}
$$

$$- i\omega \left((\tilde{\delta}_1 - \sum_{\lambda=1}^{L} \mu_\lambda \tilde{g}_\lambda^{(1)})(\Gamma_{t_0})\Gamma_{t_0}^* \right) ,$$ (32)

where a unitary element $\Gamma_t^{(i)}$ is the solution of $\frac{d}{dt}\Gamma_t^{(i)} = i\Gamma_t^{(i)}\tilde{\tau}_t^{(1)}(W_0 \otimes \mathbf{1}_1 + \kappa V)$ with $\Gamma_t^{(i)}|_{t=0} = \mathbf{1}$, $\bar{\Gamma}_{t_0} = \tilde{\tau}_{t_0}^{(1)-1}(\Gamma_{t_0})$ is again unitary, and, in deriving the first two terms, we have used a relation $\omega(\tilde{\tau}_t^{(1)}(A)) = \omega(\alpha_t^{(i)}(\tilde{\tau}_{-t}^{(1)}(A))) = \omega(\Gamma_{-t}^{(i)}A\Gamma_{-t}^{(i)*})$ resulting from the $\alpha_t^{(i)}$-invariance of ω.

Let us begin with the evaluation of the T-independent terms. Since (27) gives

$$- i(\tilde{\delta}_1 - \sum_{\lambda=1}^{L} \mu_\lambda \tilde{g}_\lambda^{(1)})(\Gamma_{t_0})\Gamma_{t_0}^* = \kappa \int_0^{t_0} ds\, \tau_s^W \left((\tilde{\delta}_1 - \sum_{\lambda=1}^{L} \mu_\lambda \tilde{g}_\lambda^{(1)})(V) \right) ,$$ (33)

the last term is evaluated as

$$\left| \omega \left((\tilde{\delta}_1 - \sum_{\lambda=1}^{L} \mu_\lambda \tilde{g}_\lambda^{(1)})(\Gamma_{t_0})\Gamma_{t_0}^* \right) \right| \leq \left\| (\tilde{\delta}_1 - \sum_{\lambda=1}^{L} \mu_\lambda \tilde{g}_\lambda^{(1)})(\Gamma_{t_0})\Gamma_{t_0}^* \right\|$$
$$\leq |\kappa| \int_0^{t_0} ds \left\| \tau_s^W \left((\tilde{\delta}_1 - \sum_{\lambda=1}^{L} \mu_\lambda \tilde{g}_\lambda^{(1)})(V) \right) \right\| = |\kappa| t_0 \left\| (\tilde{\delta}_1 - \sum_{\lambda=1}^{L} \mu_\lambda \tilde{g}_\lambda^{(1)})(V) \right\| .$$ (34)

Since $\|\Gamma_{t_0}\| = 1$ and $\|\tilde{\tau}_{t_0}^{(1)}(V)\| = \|V\|$, the third term is bounded by $|\kappa|\|V\|$:

$$\left| \omega \left(\Gamma_{t_0}\tilde{\tau}_{t_0}^{(1)}(\kappa V)\Gamma_{t_0}^* \right) \right| \leq |\kappa|\ \|V\| .$$

By taking into account the fact that $\tilde{\tau}_t^{(1)}(W(t) \otimes \mathbf{1}_1) = W(t) \otimes \mathbf{1}_1$ and comparing the equations for Γ_t and u_t, one obtains

$$\Gamma_{t_0} = u_{t_0} \otimes \mathbf{1}_1 + \Delta\Gamma_{t_0} , \qquad \Delta\Gamma_{t_0} \equiv i\kappa \int_0^{t_0} ds\, \Gamma_s \tilde{\tau}_s^{(1)}(V)((u_s^* u_{t_0}) \otimes \mathbf{1}_1) .$$ (35)

Then, because of $\tilde{\tau}_{t_0}^{(1)}((W_f - \sum_{\lambda=1}^{L} \mu_\lambda N_\lambda) \otimes \mathbf{1}_1) = (W_f - \sum_{\lambda=1}^{L} \mu_\lambda N_\lambda) \otimes \mathbf{1}_1$, the second term is evaluated as

$$\left| \omega \left(\Gamma_{t_0}\tilde{\tau}_{t_0}^{(1)}((W_f - \sum_{\lambda=1}^{L} \mu_\lambda N_\lambda) \otimes \mathbf{1}_1)\Gamma_{t_0}^* \right) - \omega \left((u_{t_0}(W_f - \sum_{\lambda=1}^{L} \mu_\lambda N_\lambda)u_{t_0}^*) \otimes \mathbf{1}_1 \right) \right|$$
$$= \left| \omega \left(\Gamma_{t_0}((W_f - \sum_{\lambda=1}^{L} \mu_\lambda N_\lambda) \otimes \mathbf{1}_1)\Gamma_{t_0}^* \right) - \omega \left((u_{t_0}(W_f - \sum_{\lambda=1}^{L} \mu_\lambda N_\lambda)u_{t_0}^*) \otimes \mathbf{1}_1 \right) \right|$$
$$\leq \left| \omega \left(\Delta\Gamma_{t_0}((W_f - \sum_{\lambda=1}^{L} \mu_\lambda N_\lambda) \otimes \mathbf{1}_1)\Gamma_{t_0}^* \right) \right| + \left| \omega \left((u_{t_0}(W_f - \sum_{\lambda=1}^{L} \mu_\lambda N_\lambda)) \otimes \mathbf{1}_1 \Delta\Gamma_{t_0}^* \right) \right|$$
$$\leq 2\|\Delta\Gamma_{t_0}\|\ \|(W_f - \sum_{\lambda=1}^{L} \mu_\lambda N_\lambda) \otimes \mathbf{1}_1\| \leq 2|\kappa|t_0\|V\|\ \|(W_f - \sum_{\lambda=1}^{L} \mu_\lambda N_\lambda) \otimes \mathbf{1}_1\| ,$$ (36)

where we have used $\|\Delta\Gamma_{t_0}\| \leq |\kappa|t_0\|V\|$.

Now we turn to the first term, which is estimated with the aid of the following lemma:

Lemma:

$$\lim_{t\to\infty} \omega(A\alpha_t^{(f)}(B)C) = \omega(AC)\omega_f(B)$$ (37)

Proof: As the KMS state ω_f is the bounded perturbation of the unique KMS state ω and $\hat{\sigma}_s^{(f)}$ commutes with $\alpha_t^{(f)}$ for all $t, s \in \mathbf{R}$, ω_f is $\alpha_t^{(f)}$-invarinat and, thus, L_f is well-defined (cf. Corollary 2.3.17 of Ref. [3]). Then, because of the assumption on the spectrum of L_f, one has, for $t \to \infty$,

$$(\psi, \pi_f(\alpha_t^{(f)}(B))\Omega_f) - (\psi, \Omega_f)(\Omega_f, \pi_f(B)\Omega_f) = (\widetilde{\psi}, e^{iL_f t}\widetilde{\varphi}_B) = \int_{-\infty}^{\infty} d\lambda e^{i\lambda t} \frac{d(\widetilde{\psi}, \hat{E}(\lambda)\widetilde{\varphi}_B)}{d\lambda} \to 0 \,,$$

where $\widetilde{\psi} = \psi - (\Omega_f, \psi)\Omega_f$ and $\widetilde{\varphi}_B = \pi_f(B)\Omega_f - (\Omega_f, \pi_f(B)\Omega_f)\Omega_f$ are absolutely continuous vectors, $\{\hat{E}(\lambda)\}$ is the spectral family of L_f and the Riemann-Lebesgue lemma is used. Corollary 5.3.9 of Ref. [3] asserts that Ω_f is separating for the bi-commutant $\pi_f(\mathcal{F})''$ of $\pi_f(\mathcal{F})$ since ω_f is a KMS state. Then, as in the proof of Theorem 4.3.23 of Ref. [3], one concludes

$$\lim_{t \to \infty} (\psi, \pi_f(\alpha_t^{(f)}(B))\varphi) = (\psi, \varphi)(\Omega_f, \pi_f(B)\Omega_f) = (\psi, \varphi)\omega_f(B) \,,$$

from the above formula. As ω is a bounded perturbation of ω_f, it is expressed by a vector $\Omega_0 \in \mathcal{H}_f$: $\omega(A) = (\Omega_0, \pi_f(A)\Omega_0)/\|\Omega_0\|^2$ (cf. Corollary 5.4.5 of Ref. [3]) and, thus, one obtains the desired result:

$$\lim_{t \to \infty} \omega(A\alpha_t^{(f)}(B)C) = \lim_{t \to \infty} (\Omega_0, \pi_f(A)\pi_f(\alpha_t^{(f)}(B))\pi_f(C)\Omega_0)/\|\Omega_0\|^2$$
$$= \omega_f(B)(\Omega_0, \pi_f(A)\pi_f(C)\Omega_0)/\|\Omega_0\|^2 = \omega(AC)\omega_f(B) \,.$$

From this lemma, one immediately obtains

$$\lim_{T \to +\infty} \omega\Big(\Gamma_{-t_0}^{(i)}\bar{\Gamma}_{t_0}\alpha_{T-t_0}^{(f)}\Big((W_f - \sum_{\lambda=1}^{L}\mu_\lambda N_\lambda)\otimes\mathbf{1}_1 + \kappa V\Big)\bar{\Gamma}_{t_0}^* \Gamma_{-t_0}^{(i)*}\Big)$$
$$= \omega_f\Big((W_f - \sum_{\lambda=1}^{L}\mu_\lambda N_\lambda)\otimes\mathbf{1}_1 + \kappa V\Big)\ \omega\Big(\Gamma_{-t_0}^{(i)}\bar{\Gamma}_{t_0}\bar{\Gamma}_{t_0}^*\Gamma_{-t_0}^{(i)*}\Big)$$
$$= \omega_f\Big((W_f - \sum_{\lambda=1}^{L}\mu_\lambda N_\lambda)\otimes\mathbf{1}_1\Big) + \omega_f(\kappa V) \,. \tag{38}$$

Moreover, $|\omega_f(\kappa V)| \le |\kappa|\|V\|$.
   In short, we have shown

$$\lim_{T \to +\infty} Q_T = \omega_f\Big((W_f - \sum_{\lambda=1}^{L}\mu_\lambda N_\lambda)\otimes\mathbf{1}_1\Big) - \omega\Big((u_{t_0}(W_f - \sum_{\lambda=1}^{L}\mu_\lambda N_\lambda)u_{t_0}^*)\otimes\mathbf{1}_1\Big) + \mathrm{O}(|\kappa|)$$

Remind that ω and ω_f are κV-perturbed KMS states of the product states $\rho_i \otimes \omega_{GC}$ and $\rho_f \otimes \omega_{GC}$, respectively, where ω_{GC} is the reservoir grand canonical state (a KMS state at β with respect to $\tilde{\tau}_t^{(1)}\tilde{\alpha}_{-\vec{\mu}t}^{(1)}$). Then, the stability of KMS states (Theorem 5.4.4 of Ref. [3]) leads to $\lim_{\kappa \to 0} \omega_f(A \otimes \mathbf{1}_1) = \mathrm{Tr}(\rho_f A)$ and $\lim_{\kappa \to 0} \omega(A \otimes \mathbf{1}_1) = \mathrm{Tr}(\rho_i A)$ ($^\forall A \in \mathcal{F}_S$, see also Ref. [25]) and, hence,

$$\lim_{\kappa \to 0}\lim_{T \to +\infty} \beta Q_T = \beta\,\mathrm{Tr}\rho_f\Big(W_f - \sum_{\lambda=1}^{L}\mu_\lambda N_\lambda\Big) - \beta\,\mathrm{Tr}\rho_i\Big(u_{t_0}(W_f - \sum_{\lambda=1}^{L}\mu_\lambda N_\lambda)u_{t_0}^*\Big)$$
$$= -\mathrm{Tr}(\rho_f \ln\rho_f) + \mathrm{Tr}\rho_i(u_{t_0}(\ln\rho_f)u_{t_0}^*) = -\mathrm{Tr}(\rho_f \ln\rho_f) + \mathrm{Tr}(\rho_{t_0}(\ln\rho_f - \ln\rho_{t_0})) + \mathrm{Tr}(\rho_i \ln\rho_i)$$
$$= S(\rho_f) - S(\rho_i) - S(\rho_f|\rho_{t_0}) \,.$$

Proof of Proposition 6: The formula (31) is still valid:

$$Q_T = -i\omega\Big(\big(\tilde{\delta}_1 - \sum_\lambda \mu_\lambda \tilde{g}_\lambda^{(1)}\big)(\Gamma_T)\Gamma_T^*\Big)$$

$$= -i\sum_{j=1}^N \left[\omega\Big(\big(\tilde{\delta}_1 - \sum_\lambda \mu_\lambda \tilde{g}_\lambda^{(1)}\big)(\Gamma_{\tilde{T}_j})\Gamma_{\tilde{T}_j}^*\Big) - \omega\Big(\big(\tilde{\delta}_1 - \sum_\lambda \mu_\lambda \tilde{g}_\lambda^{(1)}\big)(\Gamma_{\tilde{T}_{j-1}})\Gamma_{\tilde{T}_{j-1}}^*\Big)\right] . \qquad (39)$$

Let $\widetilde{\Gamma}_t^{(j)}$ be the solution of $\widetilde{\Gamma}_0^{(j)} = \mathbf{1}$, $\frac{d}{dt}\widetilde{\Gamma}_t^{(j)} = i\widetilde{\Gamma}_t^{(j)}\tilde{\tau}_t^{(1)}(W_j \otimes \mathbf{1}_1 + \kappa V)$ and $\widetilde{\Gamma}_t''^{(j)}$ be the solution of $\widetilde{\Gamma}_0''^{(j)} = \mathbf{1}$, $\frac{d}{dt}\widetilde{\Gamma}_t''^{(j)} = i\widetilde{\Gamma}_t''^{(j)}\tilde{\tau}_t^{(1)}((W_{j-1} + \varphi(t)\Delta W) \otimes \mathbf{1}_1 + \kappa V)$ with $\Delta W = (W_f - W_0)/N$, then, by comparing these equations with that of Γ_t, one has $\widetilde{\Gamma}_t^{(j)}\tilde{\tau}_t^{(1)}(A)\widetilde{\Gamma}_t^{(j)*} = \alpha_t^{(j)}(A)$, $\omega(\tilde{\tau}_t^{(1)}(A)) = \omega(\alpha_{-t}^{(0)}(\tilde{\tau}_t^{(1)}(A))) = \omega(\widetilde{\Gamma}_{-t}^{(0)}A\widetilde{\Gamma}_{-t}^{(0)*})$, $\Gamma_{\tilde{T}_j} = \Gamma_{\tilde{T}_{j-1}+t_0}\tilde{\tau}_{\tilde{T}_{j-1}+t_0}^{(1)}\big(\widetilde{\Gamma}_{T_j - t_0}^{(j)}\big)$ and $\Gamma_{\tilde{T}_{j-1}+t_0} = \Gamma_{\tilde{T}_{j-1}}\tilde{\tau}_{\tilde{T}_{j-1}}^{(1)}\big(\widetilde{\Gamma}_{t_0}''^{(j)}\big)$. Moreover,

$$-i(\tilde{\delta}_1 - \sum_\lambda \mu_\lambda \tilde{g}_\lambda^{(1)})(\widetilde{\Gamma}_t^{(j)})\widetilde{\Gamma}_t^{(j)*} = \alpha_t^{(j)}((W_j - \sum_\lambda \mu_\lambda N_\lambda) \otimes \mathbf{1}_1 + \kappa V) - (W_j - \sum_\lambda \mu_\lambda N_\lambda) \otimes \mathbf{1}_1 - \kappa V$$

Hence, we have

$$-i\omega\Big(\big(\tilde{\delta}_1 - \sum_\lambda \mu_\lambda \tilde{g}_\lambda^{(1)}\big)(\Gamma_{\tilde{T}_j})\Gamma_{\tilde{T}_j}^*\Big) + i\omega\Big(\big(\tilde{\delta}_1 - \sum_\lambda \mu_\lambda \tilde{g}_\lambda^{(1)}\big)(\Gamma_{\tilde{T}_{j-1}})\Gamma_{\tilde{T}_{j-1}}^*\Big)$$

$$= -i\omega\Big(\Gamma_{\tilde{T}_{j-1}+t_0}\tilde{\tau}_{\tilde{T}_{j-1}+t_0}^{(1)}\big(\big(\tilde{\delta}_1 - \sum_\lambda \mu_\lambda \tilde{g}_\lambda^{(1)}\big)(\widetilde{\Gamma}_{T_j-t_0}^{(j)})\widetilde{\Gamma}_{T_j-t_0}^{(j)*}\big)\Gamma_{\tilde{T}_{j-1}+t_0}^*\Big)$$

$$\quad - i\omega\Big(\big(\tilde{\delta}_1 - \sum_\lambda \mu_\lambda \tilde{g}_\lambda^{(1)}\big)(\Gamma_{\tilde{T}_{j-1}+t_0})\Gamma_{\tilde{T}_{j-1}+t_0}^*\Big) + i\omega\Big(\big(\tilde{\delta}_1 - \sum_\lambda \mu_\lambda \tilde{g}_\lambda^{(1)}\big)(\Gamma_{\tilde{T}_{j-1}})\Gamma_{\tilde{T}_{j-1}}^*\Big)$$

$$= -i\omega\Big(\Gamma_{\tilde{T}_{j-1}+t_0}\tilde{\tau}_{\tilde{T}_{j-1}+t_0}^{(1)}\big(\big(\tilde{\delta}_1 - \sum_\lambda \mu_\lambda \tilde{g}_\lambda^{(1)}\big)(\widetilde{\Gamma}_{T_j-t_0}^{(j)})\widetilde{\Gamma}_{T_j-t_0}^{(j)*}\big)\Gamma_{\tilde{T}_{j-1}+t_0}^*\Big)$$

$$\quad - i\omega\Big(\Gamma_{\tilde{T}_{j-1}}\tilde{\tau}_{\tilde{T}_{j-1}}^{(1)}\big(\big(\tilde{\delta}_1 - \sum_\lambda \mu_\lambda \tilde{g}_\lambda^{(1)}\big)(\widetilde{\Gamma}_{t_0}''^{(j)})\big)\Gamma_{\tilde{T}_{j-1}+t_0}^*\Big)$$

$$= \omega\Big(\Gamma_{\tilde{T}_{j-1}+t_0}\tilde{\tau}_{\tilde{T}_{j-1}+t_0}^{(1)}\big(\alpha_{T_j-t_0}^{(j)}\big((W_j - \sum_\lambda \mu_\lambda N_\lambda) \otimes \mathbf{1}_1 + \kappa V\big)\big)\Gamma_{\tilde{T}_{j-1}+t_0}^*\Big)$$

$$\quad - \omega\Big(\Gamma_{\tilde{T}_{j-1}+t_0}\tilde{\tau}_{\tilde{T}_{j-1}+t_0}^{(1)}\big((W_j - \sum_\lambda \mu_\lambda N_\lambda) \otimes \mathbf{1}_1 + \kappa V\big)\Gamma_{\tilde{T}_{j-1}+t_0}^*\Big)$$

$$\quad - i\omega\Big(\Gamma_{\tilde{T}_{j-1}}\tilde{\tau}_{\tilde{T}_{j-1}}^{(1)}\big(\big(\tilde{\delta}_1 - \sum_\lambda \mu_\lambda \tilde{g}_\lambda^{(1)}\big)(\widetilde{\Gamma}_{t_0}''^{(j)})\widetilde{\Gamma}_{t_0}''^{(j)*}\big)\Gamma_{\tilde{T}_{j-1}}^*\Big)$$

$$= \omega\Big(\widetilde{\Gamma}_{-\tilde{T}_{j-1}-t_0}^{(0)}\bar{\Gamma}_{\tilde{T}_{j-1}+t_0}\big\{\alpha_{T_j-t_0}^{(j)}\big((W_j - \sum_\lambda \mu_\lambda N_\lambda) \otimes \mathbf{1}_1 + \kappa V\big)\big\}\bar{\Gamma}_{\tilde{T}_{j-1}+t_0}^*\widetilde{\Gamma}_{-\tilde{T}_{j-1}-t_0}^{(0)*}\Big)$$

$$\quad - \omega\Big(\Gamma_{\tilde{T}_{j-1}}\tilde{\tau}_{\tilde{T}_{j-1}}^{(1)}\big(\widetilde{\Gamma}_{t_0}''^{(j)}\tilde{\tau}_{t_0}^{(1)}\big((W_j - \sum_\lambda \mu_\lambda N_\lambda) \otimes \mathbf{1}_1 + \kappa V\big)\widetilde{\Gamma}_{t_0}''^{(j)*}\big)\Gamma_{\tilde{T}_{j-1}}^*\Big)$$

$$\quad - i\omega\Big(\Gamma_{\tilde{T}_{j-1}}\tilde{\tau}_{\tilde{T}_{j-1}}^{(1)}\big(\big(\tilde{\delta}_1 - \sum_\lambda \mu_\lambda \tilde{g}_\lambda^{(1)}\big)(\widetilde{\Gamma}_{t_0}''^{(j)})\widetilde{\Gamma}_{t_0}''^{(j)*}\big)\Gamma_{\tilde{T}_{j-1}}^*\Big) ,$$

where $\bar{\Gamma}_t \equiv \tilde{\tau}_t^{(1)-1}(\Gamma_t)$. On the other hand, the states ω, ω_j and the evolutions $\alpha_t^{(j)}$ $(j = 1, 2, \cdots N)$ satisfy the assumptions necessary for the Lemma, we have

$$\lim_{T_j \to +\infty} \omega\Big(\widetilde{\Gamma}_{-\tilde{T}_{j-1}-t_0}^{(0)}\bar{\Gamma}_{\tilde{T}_{j-1}+t_0}\alpha_{T_j-t_0}^{(j)}\big(A\big)\bar{\Gamma}_{\tilde{T}_{j-1}+t_0}^*\widetilde{\Gamma}_{-\tilde{T}_{j-1}-t_0}^{(0)*}\Big)$$

$$= \omega\Big(\widetilde{\Gamma}_{-\tilde{T}_{j-1}-t_0}^{(0)}\bar{\Gamma}_{\tilde{T}_{j-1}+t_0}\bar{\Gamma}_{\tilde{T}_{j-1}+t_0}^*\widetilde{\Gamma}_{-\tilde{T}_{j-1}-t_0}^{(0)*}\Big)\omega_j(A) = \omega_j(A) , \qquad (40)$$

$$\lim_{T_{j-1} \to +\infty} \omega\Big(\Gamma_{\tilde{T}_{j-1}}\tilde{\tau}_{\tilde{T}_{j-1}}^{(1)}(A)\Gamma_{\tilde{T}_{j-1}}^*\Big) = \lim_{T_{j-1} \to +\infty} \omega\Big(\Gamma_{\tilde{T}_{j-2}+t_0}\tilde{\tau}_{\tilde{T}_{j-2}+t_0}^{(1)}\big(\alpha_{T_{j-1}-t_0}^{(j)}(A)\big)\Gamma_{\tilde{T}_{j-2}+t_0}^*\Big)$$

$$= \omega\Big(\widetilde{\Gamma}_{-\tilde{T}_{j-1}-t_0}^{(0)}\bar{\Gamma}_{\tilde{T}_{j-2}+t_0}\bar{\Gamma}_{\tilde{T}_{j-2}+t_0}^*\widetilde{\Gamma}_{-\tilde{T}_{j-2}-t_0}^{(0)*}\Big)\omega_{j-1}(A) = \omega_{j-1}(A) . \qquad (41)$$

Therefore, by taking the limits $T_j \to +\infty$, $T_{j-1} \to +\infty$ in this order, we get

$$\lim_{T_{j-1}\to\infty}\lim_{T_j\to\infty}\left[-i\omega\Big((\tilde\delta_1-\sum_\lambda\mu_\lambda\tilde g_\lambda^{(1)})(\Gamma_{\tilde T_j})\Gamma^*_{\tilde T_j}\Big)+i\omega\Big((\tilde\delta_1-\sum_\lambda\mu_\lambda\tilde g_\lambda^{(1)})(\Gamma_{\tilde T_{j-1}})\Gamma^*_{\tilde T_{j-1}}\Big)\right]$$

$$=\omega_j\Big((W_j-\sum_\lambda\mu_\lambda N_\lambda)\otimes\mathbf{1}_1+\kappa V\Big)-\omega_{j-1}\Big(\widetilde\Gamma_{t_0}''^{(j)}\widetilde\tau_{t_0}^{(1)}\Big((W_j-\sum_\lambda\mu_\lambda N_\lambda)\otimes\mathbf{1}_1+\kappa V\Big)\widetilde\Gamma_{t_0}''^{(j)*}\Big)$$

$$-i\omega_{j-1}\Big((\tilde\delta_1-\sum_\lambda\mu_\lambda\tilde g_\lambda^{(1)})(\widetilde\Gamma_{t_0}''^{(j)})\widetilde\Gamma_{t_0}''^{(j)*}\Big)\ .$$

As before, one has

$$\max\left\{|\omega_j(\kappa V)|,\Big|\omega_{j-1}(\widetilde\Gamma_{t_0}''^{(j)}\widetilde\tau_{t_0}^{(1)}(\kappa V)\widetilde\Gamma_{t_0}''^{(j)*})\Big|\right\}\le|\kappa|\ \|V\|\ ,\tag{42}$$

$$\left|\omega_{j-1}(\tilde\delta_1-\sum_\lambda\mu_\lambda\tilde g_\lambda^{(1)})(\widetilde\Gamma_{t_0}''^{(j)})\widetilde\Gamma_{t_0}''^{(j)*}\right|\le|\kappa|\ t_0\ \|(\tilde\delta_1-\sum_\lambda\mu_\lambda\tilde g_\lambda^{(1)})(V)\|\ ,\tag{43}$$

$$\left|\omega_{j-1}\Big(\widetilde\Gamma_{t_0}''^{(j)}\widetilde\tau_{t_0}^{(1)}\Big((W_j-\sum_\lambda\mu_\lambda N_\lambda)\otimes\mathbf{1}_1\Big)\widetilde\Gamma_{t_0}''^{(j)*}\Big)-\omega_{j-1}\Big([\tilde u_{t_0}^{(j)}(W_j-\sum_\lambda\mu_\lambda N_\lambda)\tilde u_{t_0}^{(j)*}]\otimes\mathbf{1}_1\Big)\right|$$

$$\le2|\kappa|\ t_0\ \|V\|\ \Big\|(W_j-\sum_\lambda\mu_\lambda N_\lambda)\otimes\mathbf{1}_1\Big\|\ ,\tag{44}$$

where $\tilde u_t^{(j)}$ is the solution of $\tilde u_0^{(j)}=\mathbf{1}_S$, $\frac{d}{dt}\tilde u_t^{(j)}=i\tilde u_t^{(j)}(W_{j-1}+\varphi(t)\Delta W)$. Moreover, the state ω_j is a κV-perturbed KMS state of the product state $\rho_j\otimes\omega_{GC}$, where $\rho_j=\exp(-\beta(W_j-\sum_\lambda\mu_\lambda N_\lambda))/\Xi_j$ with Ξ_j the grand partition function, and, thus, $\lim_{\kappa\to0}\omega_j(A\otimes\mathbf{1}_1)=\mathrm{Tr}(A\rho_j)$ ($\forall A\in\mathcal F_S$). As a consequence, one gets

$$\lim_{\kappa\to0}\lim_{T_{j-1}\to\infty}\lim_{T_j\to\infty}\left[-i\omega\left((\tilde\delta_1-\sum_\lambda\mu_\lambda\tilde g_\lambda^{(1)})(\Gamma_{\tilde T_j})\Gamma^*_{\tilde T_j}\right)+i\omega\left((\tilde\delta_1-\sum_\lambda\mu_\lambda\tilde g_\lambda^{(1)})(\Gamma_{\tilde T_{j-1}})\Gamma^*_{\tilde T_{j-1}}\right)\right]$$

$$=\mathrm{Tr}\Big(\rho_j(W_j-\sum_\lambda\mu_\lambda N_\lambda)\Big)-\mathrm{Tr}\Big(\rho_{j-1}(\tilde u_{t_0}^{(j)}(W_j-\sum_\lambda\mu_\lambda N_\lambda)\tilde u_{t_0}^{(j)*})\Big)$$

$$=\beta^{-1}\left\{-\mathrm{Tr}(\rho_j\ln\rho_j)+\mathrm{Tr}\Big(\tilde\rho_{j-1}\ln\rho_j\Big)\right\}=\beta^{-1}\left\{S(\rho_j)-S(\rho_{j-1})-S(\rho_j|\tilde\rho_{j-1})\right\}\ .$$

where $\tilde\rho_{j-1}=\tilde u_{t_0}^{(j)*}\rho_{j-1}\tilde u_{t_0}^{(j)}$, and, thus,

$$\lim_{\kappa\to0}\lim_{T_1\to+\infty}\lim_{T_2\to+\infty}\cdots\lim_{T_N\to+\infty}\beta Q_T=S(\rho_f)-S(\rho_i)-\sum_{j=1}^N S(\rho_j|\tilde\rho_{j-1})\ ,$$

where $\rho_i\equiv\rho_0$ and $\rho_f\equiv\rho_N$.

Now we investigate the contribution from the relative entropies. Reminding that N_λ commutes with W_0 and W_f, one finds

$$S(\rho_j|\tilde\rho_{j-1})=\beta\langle\Delta\tilde u_{t_0}^{(j)}W\tilde u_{t_0}^{(j)*}\rangle_{j-1}+\beta(\langle\tilde u_{t_0}^{(j)}W_{j-1}\tilde u_{t_0}^{(j)*}\rangle_{j-1}-\langle W_{j-1}\rangle_{j-1})+\ln\langle e^{\beta W_{j-1}}e^{-\beta W_j}\rangle_{j-1}\ ,$$

where $\langle A\rangle_{j-1}\equiv\mathrm{Tr}(\rho_{j-1}A)$ ($\forall A\in\mathcal F_S$). On the other hand, we have

$$\tilde u_{t_0}^{(j)}\Delta W\tilde u_{t_0}^{(j)*}=e^{iW_{j-1}t_0}\Delta We^{-iW_{j-1}t_0}+e^{iW_{j-1}t_0}\Delta W\ \Delta\tilde u_{t_0}^{(j)*}+\Delta\tilde u_{t_0}^{(j)}\ \Delta W\tilde u_{t_0}^{(j)*}\ ,$$

$$\tilde u_{t_0}^{(j)}W_{j-1}\tilde u_{t_0}^{(j)*}-W_{j-1}=i\int_0^{t_0}ds\varphi(s)e^{iW_{j-1}s}[\Delta W,W_{j-1}]e^{-iW_{j-1}s}$$

$$+i\int_0^{t_0}ds\varphi(s)\left\{e^{iW_{j-1}s}[\Delta W,W_{j-1}]\ \Delta\tilde u_s^{(j)*}+\Delta\tilde u_s^{(j)}\ [\Delta W,W_{j-1}]\tilde u_s^{(j)*}\right\}\ ,$$

$$e^{\beta W_{j-1}}e^{-\beta W_j}=\mathbf{1}_S-\int_0^\beta d\tau e^{\tau W_{j-1}}\Delta We^{-\tau W_{j-1}}$$

$$+ \int_0^\beta d\tau \int_0^\tau d\tau' e^{\tau W_{j-1}} \Delta W e^{-(\tau-\tau')W_{j-1}} \Delta W e^{-\tau' W_j} \; ,$$

where $\Delta \widetilde{u}_{t_0}^{(j)} = i \int_0^{t_0} ds \varphi(s) \widetilde{u}_s^{(j)} \Delta W e^{iW_{j-1}(t_0-s)}$, and, since ρ_{j-1} commutes with W_{j-1} and $\|W_j\| \leq \|W_0\| + j/N\|W_f - W_0\| \leq \|W_0\| + \|W_f - W_0\| \equiv K$,

$$\left| \langle \widetilde{u}_{t_0}^{(j)} \Delta W \widetilde{u}_{t_0}^{(j)*} \rangle_{j-1} - \langle \Delta W \rangle_{j-1} \right| \leq 2\|\Delta W\|^2 \int_0^{t_0} ds |\varphi(s)| \; ,$$

$$\left| \langle \widetilde{u}_{t_0}^{(j)} W_{j-1} \widetilde{u}_{t_0}^{(j)*} \rangle_{j-1} - \langle W_{j-1} \rangle_{j-1} \right| \leq 4K\|\Delta W\|^2 \int_0^{t_0} ds \int_0^s ds' |\varphi(s)\varphi(s')| \; ,$$

$$\left| \langle e^{\beta W_{j-1}} e^{-\beta W_j} \rangle_{j-1} - 1 + \beta \langle \Delta W \rangle_{j-1} \right| \leq \|\Delta W\|^2 \int_0^\beta d\tau \int_0^\tau d\tau' e^{K(\tau+\tau'+|\tau-\tau'|)} \; ,$$

where we have used $\langle [\Delta W, W_{j-1}] \rangle_{j-1} = 0$. Therefore, if N is large enough, we obtain $|S(\rho_j | \widetilde{\rho}_{j-1})| \leq K'\|\Delta W\|^2 = K'\|W_f - W_0\|^2 / N^2$ with some positive constant K', and the desired result:

$$\left| \lim_{\kappa \to 0} \lim_{T_1 \to +\infty} \lim_{T_2 \to +\infty} \cdots \lim_{T_N \to +\infty} \beta Q_T - \{S(\rho_f) - S(\rho_i)\} \right| \leq \frac{K'\|W_f - W_0\|^2}{N} \; .$$

V. CONCLUSION

In the first half of this article, we have shown that, if the evolution is L^1-asymptotic abelian and the Møller morphism relating a nonequilibrium steady state to a local equilibrium state is invertible, the natural non-equilibrium steady states can be regarded as MacLennan-Zubarev nonequilibrium ensembles. At first sight, invertibility of the Møller morphisms seems to be too strong since one can easily find a counter example. However, for several systems, the division of the whole system without a finite part (i.e. the case where $\mathcal{F}_S = \emptyset$) provides invertible Møller morphisms and, thus, we believe that Proposition 2 holds generically provided that the system is divided appropriately. Also it should be emphasized that Proposition 2 does not exclude the possibility that the class of natural nonequilibrium steady states is wider than that of MacLennan-Zubarev ensembles since the generator of the automorphisims defining the steady states takes MacLennan-Zubarev form only in a subset of its domain.

In the second half, we have shown that the small system coupled with a single reservoir would follow 'thermodynamic' processes and satisfies the Clausius inequality/equality. However, it should not be regarded as a dynamical proof of the second law of thermodynamics since we start from canonical ensembles which are very outcome of the second law. We think that the importance of this observation lies in the facts that (i) the dynamical evolution is consistent with thermodynamics, (ii) one can define thermodynamic heat microscopically, (iii) entropy generation is identified for step-wise evolution, and (iv) a characterization of a quasistatic change is given.

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[30] Throughout this article, the subalgebra $\mathbf{1}_S \otimes \mathbf{1}_1 \otimes \cdots \otimes \mathcal{F}_k \otimes \cdots \otimes \mathbf{1}_M$, where $\mathbf{1}_S$ and $\mathbf{1}_j$ are unities, respectively, of $\mathcal{F}_S$ and $\mathcal{F}_j$, is abbrebiated as $\mathcal{F}_j$. Similarly, $\mathcal{F}_S \otimes \mathbf{1}_1 \otimes \cdots \otimes \mathbf{1}_M$ as $\mathcal{F}_S$.

[31] Namely, there exist a Hilbert space $\mathcal{H}_f$ and a *-morphism π_f from $\mathcal{F}$ into the algebra

of all bounded operators on $\mathcal{H}_f$ and a unit vector $\Omega_f \in \mathcal{H}_f$, such that the closure of $\pi_f(\mathcal{F})\Omega_f$ is equal to $\mathcal{H}_f$.

[32] The reference state is defined as an equilibrium state with instantaneous parameter values.